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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
YOSHIMASA TANAKA, ET AL. : EXAMINER: HESS, BRUCE H
SERIAL NO: 10/566,202 :
FILED: JANUARY 27, 2006 : GROUP ART UNIT: 1794
FOR: THERMAL TRANSFER :
RECEIVING SHEET, PRODUCTION
METHOD THEREOF AND IMAGE
FORMING METHOD USING THE
SHEET

DECLARATION UNDER 37 C.F.R. 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

I, Masato Kawamura hereby declare:

1. I am a named inventor of the above-identified application and am familiar with the specification of the above-identified patent application.

2. The following observations and experiments were carried out by me or under my direct supervision and control.

3. In order to show the remarkable and unexpected results of use of two kinds of hollows particles satisfying in the requirement specified in claim 1 of the present

application in the thermal transfer receiving sheet, we had performed direct comparative experiments which are shown below. These experiments demonstrate use of the two kinds of hollow particles in the receiving layer of a thermal transfer sheet is significantly advantageous in preventing generation of dents, spike marks or the like on its surface.

4. Experiments

Example A

[Formation of Intermediate Layer]

Using a 150 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 174.4 g/m^2) as the sheet-like support, Intermediate Layer Coating Solution 1 having the following composition was coated to have a dry thickness of 48 μm and dried to form an intermediate layer.

Intermediate Layer Coating Solution 1

Hollow particle A: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 3.8 μm, coefficient of variation in particle diameter: 14%, hollow percentage by volume: 75%)	65 parts

Hollow particle B: microcapsule-type hollow particle (Nipol MH-5055, trade name, produced by ZEON Corporation, average particle diameter: 0.55 μm, coefficient of variation in particle diameter: 15%, hollow percentage by volume: 55%)	0.5 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	24.5 parts
Water	200 parts

[Formation of Receiving Sheet]

On the intermediate layer, Barrier Layer Coating Solution 1 having the following composition was coated to have a coated amount in terms of solid content of 2 g/m^2 and dried to form a barrier layer. Subsequently, on the barrier layer, Receiving Layer Coating Solution 1 having the following composition was coated to have a coated amount in terms of solid content of 5 g/m^2 and dried. The formed coating was then cured at 50°C for 72 hours to form a receiving layer. In this way, a receiving sheet was prepared. Thereafter, on the sheet-like support surface opposite the side where the receiving layer was provided, Back Surface Layer Coating Solution 1 having the following composition was coated to have a coated amount in terms of solid content of 3 g/m^2 and dried, thereby obtaining a receiving sheet.

Barrier Layer Coating Solution 1

Swelling inorganic layered compound (sodium tetrasilicon mica, average particle long diameter: 6.3 μm, aspect ratio: 2,700)	30 parts
Polyvinyl alcohol (PVA105, trade name, produced by Kuraray Co., Ltd.)	50 parts
Styrene-butadiene latex (L-1537, trade name, produced by Asahi Kasei Corporation)	20 parts
Water	1,100 parts

Receiving Layer Coating Solution 1

Polyester resin (VYLON 200, trade name, produced by Toyobo Co., Ltd.)	100 parts
Silicone oil (KF393, trade name, produced by Shin-Etsu Chemical Co., Ltd.)	3 parts
Polyisocyanate (TAKENATE D-140N, trade name, produced by Takeda Chemical Industries, Ltd.)	5 parts
A 1/1 (by mass) mixed solution of toluene/methyl ethyl ketone	400 parts

Back Surface Layer Coating Solution 1

Polyvinyl acetal resin (ESLEC KX-1, produced by Sekisui Chemical Co., Ltd.)	40 parts
Polyacrylic acid ester resin (JURYMER AT613, trade name, produced by Nihon Junyaku Co., Ltd.)	20 parts

Nylon resin particle (MW330, trade name, produced by Shinto Fine Co., Ltd.)	10 parts
Zinc stearate (Z-7-30, trade name, produced by Chukyo Yushi Co., Ltd.)	10 parts
Cationic electrically conducting resin (CHEMISTAT 9800, trade name, produced by Sanyo Chemical Industries Co., Ltd.)	10 parts
A 2/3 (by mass) mixed solution of water/isopropyl alcohol	400 parts

Example B

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that in the formation of the intermediate layer, Intermediate Layer Coating Solution 2 having the following composition was used.

<u>Intermediate Layer Coating Solution 2</u>

Hollow particle A: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 3.8 μm, coefficient of variation in particle diameter: 14%, hollow percentage by volume: 75%)	65 parts

Hollow particle B: microcapsule-type hollow particle (ROHPAKE HP-1055, trade name, produced by Rohm & Haas, average particle diameter: 1.0 μm, coefficient of variation in particle diameter: 12%, hollow percentage by volume: 55%)	3 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	22 parts
Water	200 parts

Example C

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 105 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 127.9 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 3 having the following composition was used.

Intermediate Layer Coating Solution 3

Hollow particle A: prefoamed hollow particle (EXPANCEL 551DE20, trade name, produced by Japan Fillite Co.Ltd., average particle diameter: 20 μm, coefficient of variation in particle diameter: 5%, hollow percentage by volume: 94%)	40 parts

Hollow particle B: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 3.8 μm, coefficient of variation in particle diameter: 14%, hollow percentage by volume: 75%)	1.5 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	48.5 parts
Water	200 parts

Example D

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 105 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 127.9 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 4 having the following composition was coated to have a dry thickness of 100 μm and dried to form an intermediate layer.

Intermediate Layer Coating Solution 4

Hollow particle A: prefoamed hollow particle (EXPANCEL 551DE20, trade name, produced by Japan Fillite Co.Ltd., average particle diameter: 20 µm, coefficient of variation in particle diameter: 5%, hollow percentage by volume: 94%)	40 parts
Hollow particle B: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 5.4 µm, coefficient of variation in particle diameter: 10%, hollow percentage by volume: 80%)	3.5 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	46.5 parts
Water	200 parts

Example E

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 105 µm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 127.9 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 5 having the following composition was coated to have a dry thickness of 93 µm and dried to form an intermediate layer.

Intermediate Layer Coating Solution 5

Hollow particle A: prefoamed complexed hollow particle (main component of the shell: polyacrylonitrile, shell surface being coated with calcium carbonate powder, average particle diameter: 20 μm, coefficient of variation in particle diameter: 5%, hollow percentage by volume: 90%, particle density: 0.2g/cm^3)	45 parts
Hollow particle B: microcapsule-type hollow particle (Honen Microsphere MB-925, trade name, produced by Honen Corporation, average particle diameter: 5 μm, coefficient of variation in particle diameter: 11%, hollow percentage by volume: 35%)	3.5 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	41.5 parts
Water	200 parts

Comparative Example 1

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 105 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 127.9 g/m^2) was used as the sheet-like support and in

the formation of the intermediate layer, Intermediate Layer Coating Solution 6 having the following composition was coated to have a dry thickness of 108 μm and dried to form an intermediate layer.

Intermediate Layer Coating Solution 5

Hollow particle A: prefoamed hollow particle (EXPANCEL 091DE, trade name, produced by Japan Fillite Co.Ltd., average particle diameter: 40 μm, coefficient of variation in particle diameter: 4%, hollow percentage by volume: 97%)	30 parts
Hollow particle B: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 3.8 μm, coefficient of variation in particle diameter: 14%, hollow percentage by volume: 75%)	0.3 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	59.7 parts
Water	200 parts

Comparative Example 2

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 150 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co.,

Ltd., 174.4 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 6 having the following composition was coated to have a dry thickness of 51 μm and dried to form an intermediate layer.

<u>Intermediate Layer Coating Solution 7</u>

Hollow particle A: microcapsule-type hollow particle (ROHPAKE HP-1055, trade name, produced by Rohm & Haas, average particle diameter: 1.0 μm, coefficient of variation in particle diameter: 12%, hollow percentage by volume: 55%)	60 parts
Hollow particle B: microcapsule-type hollow particle (SX863A, trade name, produced by JSR, average particle diameter: 0.4 μm, coefficient of variation in particle diameter: 13%, hollow percentage by volume: 30%)	8 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	22 parts
Water	200 parts

Comparative Example 3

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 105 μm-thick art

paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 127.9 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 8 having the following composition was coated to have a dry thickness of 93 μm and dried to form an intermediate layer.

<u>Intermediate Layer Coating Solution 8</u>

Hollow particle A: prefoamed hollow particle (EXPANCEL 551DE20, trade name, produced by Japan Fillite Co.Ltd., average particle diameter: 20 μm, coefficient of variation in particle diameter: 5%, hollow percentage by volume: 94%)	40 parts
Hollow particle B: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 10 μm, coefficient of variation in particle diameter: 10%, hollow percentage by volume: 80%)	22 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	28 parts
Water	200 parts

Comparative Example 4

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the

same manner as in Example A except that a 105 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 127.9 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 9 having the following composition was coated to have a dry thickness of 95 μm and dried to form an intermediate layer.

<u>Intermediate Layer Coating Solution 9</u>

Hollow particle A: prefoamed hollow particle (EXPANCEL 461DE, trade name, produced by Japan Fillite Co.Ltd., average particle diameter: 30 μm, coefficient of variation in particle diameter: 4%, hollow percentage by volume: 94%)	30 parts
Hollow particle B: microcapsule-type hollow particle (ROHPAKE HP-1055, trade name, produced by Rohm & Haas, average particle diameter: 1.0 μm, coefficient of variation in particle diameter: 12%, hollow percentage by volume: 55%)	0.01 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	60 parts
Water	200 parts

Comparative Example 5

A receiving sheet was prepared and the receiving sheet after printing was subjected to a pressure treatment in the same manner as in Example A except that a 150 μm-thick art paper (OK Kinfuji N, trade name, produced by Oji Paper Co., Ltd., 174.4 g/m^2) was used as the sheet-like support and in the formation of the intermediate layer, Intermediate Layer Coating Solution 10 having the following composition was coated to have a dry thickness of 49 μm and dried to form an intermediate layer.

Intermediate Layer Coating Solution 10

Hollow particle A: prefoamed hollow particle mainly comprising polyacrylonitrile (average particle diameter: 3.8 μm, coefficient of variation in particle diameter: 14%, hollow percentage by volume: 75%)	65 parts
Polyvinyl alcohol (PVA205, trade name, produced by Kuraray Co., Ltd.)	10 parts
Styrene-butadiene latex (PT1004, trade name, produced by ZEON Corporation)	25 parts
Water	200 parts

Evaluation

The receiving sheets obtained in Examples above were evaluated by the following methods and the results obtained are shown in the Table.

[Printing Quality] (printing density, image uniformity)

Using a commercially available thermal transfer video

printer (UP-DR100, trade name, manufactured by Sony Corp.), ink layers for three colors of an ink sheet comprising a 6 μm-thick polyester film having provided thereon ink layers each comprising a sublimable dye of yellow, magenta or cyan and a binder were sequentially contacted with the receiving sheet for the test and subjected to heating stepwise controlled by a thermal head to thermally transfer a predetermined image to the receiving sheet, whereby a halftone monochromatic image of each color or a color mixed image was printed.

The reflection density of the recorded image transferred to the receiving sheet was measured by a Macbeth reflection densitometer (RD-914, trade name, manufactured by Kollmorgen) with respect to each applied energy. The reflection density in the high gradation part corresponding to the 15th step from the lower side of the applied energy is shown as the printing density in Table 1. When the printing density is 2.0 or more, the receiving sheet is sufficiently suited for practical use.

Furthermore, the uniformity of the recorded image in the gradation portion corresponding to an optical density (black) of 0.3 was evaluated by observing with an eye whether irregular shading and white spot were present or not. The evaluation results were rated ◎ when excellent, ○ when good, △ when irregular shading and white spot were observed, or × when defects of irregular shading and white spot were serious.

When the evaluation is the ○ level or higher, the receiving sheet is sufficiently suited for practical use.

[Dents on Receiving Sheet]

A commercially available thermal transfer video printer (M1, trade name, manufactured by Sony Corp.) was modified to increase the nip pressure of its transfer roller. The nip pressure was measured to be 50kg/cm^2 by use of a film for testing pressure (Prescale, trade name, manufactured by Fuji Film Corp.). By use of this testing system, generation of dents in the receiving sheet caused by the transfer roller was visually evaluated by naked eye.

The evaluation results were rated ◎ when no generation of dents was visible, ○ when generation of dents was rarely visible, or × when significant generation of dents was visible.

When the evaluation is the ○ level or higher, the receiving sheet is sufficiently suited for practical use.

Table

	Prefoamed Hollow Particle A				Prefoamed Hollow Particle B				L_B/L_A	Printing Density	Image Uniformity	Dents in Receiving Sheet
	Average Particle Diameter L_A (μm)	Coefficient of Variation in Particle Diameter (%)	Hollow Percentage by Volume (%)	Parts by Weight	Average Particle Diameter L_B (μm)	Coefficient of Variation in Particle Diameter (%)	Hollow Percentage by Volume (%)	Parts by Weight				
Ex. A	3.8	14	75	65	0.55	15	55	0.5	0.14	2.11	◎	○
Ex. B	3.8	14	75	65	1	12	55	3	0.26	2.11	◎	◎
Ex. C	20	5	94	40	3.8	14	75	1.5	0.19	2.16	○	○
Ex. D	20	5	94	40	5.4	10	80	3.5	0.27	2.15	○	◎
Ex. E	20	5	90	45	5	11	35	3.5	0.25	2.08	○	◎
Comp. Ex. 1	40	4	97	30	3.8	14	75	0.3	0.10	2.15	×	×
Comp. Ex. 2	1	12	55	60	0.4	13	30	8	0.40	1.91	○	○
Comp. Ex. 3	20	5	94	40	10	10	80	22	0.50	1.93	×	×
Comp. Ex. 4	30	4	94	30	1	12	55	0.01	0.03	2.11	×	×
Comp. Ex. 5	3.8	14	75	65	-	-	-	-	-	2.03	○	×

5. Examples A to D show that use of two kinds of hollow particles satisfying the requirements prescribed in current claim 1 provides thermal transfer sheet with excellent printing density, image uniformity and reduced number of dents.

Comparative Example 1 shows that use of two kinds of hollow particles, one of which has an average particle diameter larger than 35μm, is unfavorable for image uniformity and dents on the surface of the receiving sheet.

Comparative Example 2 shows that use of two kinds of hollow particles, one of which has an average particle diameter smaller than 2μm, is unfavorable for printing density of the receiving sheet.

Comparative Example 3 shows that use of two kinds of hollow particles, the ratio of the average particle diameters (LB/LA) is larger than 0.4, is unfavorable for printing density, image uniformity and dents on the surface of the receiving sheet.

Comparative Example 4 shows that use of two kinds of hollow particles, the ratio of the average particle diameters (LB/LA) is less than 0.05, is unfavorable for image uniformity and dents on the surface of the receiving sheet of the receiving sheet.

Comparative Example 5 shows that use of only one kind of hollow particles is unfavorable for printing density of the receiving sheet.

None of the references (JP'651, JP943, JP'968) describe or suggest the remarkable and unexpected effects of use of the two kinds of hollow particles.

Therefore, the present invention provides a remarkable

effect over the disclosures in the references.

6. The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believe to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Masato Kawamura

Masato Kawamura

February 25, 2009

Date